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Flux-Gradient Estimates of Ammonia Emissions from Beef Cattle Feedyard Pens

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Abstract. Concentrated animal feeding operations are major sources of ammonia emitted to the atmosphere. There is a considerable literature on ammonia emissions from poultry and swine, but few studies have investigated large, open lot beef cattle feedyards. We used the micrometeorological flux-gradient method to estimate ammonia emissions during six field campaigns in three seasons. Profiles of ammonia, wind speed and air temperature were measured on towers located within feedyard pens. Atmospheric ammonia concentration was measured using either acid gas washing or chemiluminescence. Mean daily ammonia flux in summer averaged 72 µg m^{-2} s^{-1}, and in winter, 39 µg m^{-2} s^{-1}. Springtime fluxes were highly variable and averaged 79 µg m^{-2} s^{-1}; high springtime fluxes were attributed to greater ammonium concentration in manure and high wind speeds. Ammonia-N emisson rate averaged 3930 and 2150 kg d^{-1} in summer and winter, respectively, which was 45% and 27% of fed N. Assuming that the mean of summer and winter emission rates represented a mean annual emission, ammonia-N loss was 36% of fed N, and an annual emission factor for feedyard pens based on total yearly per head production was 11.0 kg NH_3-N $head^{-1}$ yr^{-1}. Ammonia emissions increased after N in cattle rations was increased from 13.5% to 14.5%. Ammonia emissions estimated using the flux-gradient method were 22 to 36% less than those derived from an inverse dispersion model. Uncertainty in the Schmidt number and possible violation of the assumption of homogeneous flow could have contributed to the lower flux estimates of the flux-gradient method. Optimizing fed N through practices such as phase feeding could help minimize ammonia emissions. Longer term, more continuous monitoring of ammonia emissions is needed to better define annual variability, emission rates and factors, and facilitate development of process models.

Introduction

Human activities have doubled the nitrogen that cycles through terrestrial ecosystems (Smil, 1990; Vitousek et al., 1997). These include the production and use of nitrogen fertilizers, planting of nitrogen-fixing crops, release of nitrogen from storage reservoirs in soils and plant biomass, and the burning of fossil fuels. Many of these sources are agricultural, and estimates of the agricultural contribution to increased N in global ecosystems range from 50 to more than 90%, with animal husbandry comprising the majority of that contribution (Bouwman et al., 1997; Ferm, 1998; Galloway and Cowling, 2002; Howarth et al., 2002).

Ammonia is the primary basic constituent of the atmosphere; it readily combines with and neutralizes oxidized compounds such as SO_2 and NO_x, forming secondary particulates. Ammonia and its compounds can be transported long distances, wash out in precipitation, and return to the earth's surface. Nitrogen enrichment of land and water can have varying effects, from the fertilization of crop land to excessive enrichment and degradation of sensitive ecosystems (Matson et al., 2002; Rabalais, 2002; Todd et al., 2004). Animal feeding operations, with abundant manure, are major sources of ammonia. Ammonia emissions from beef cattle feedyards have not been well-researched, compared with other species, such as swine and poultry.

Micrometeorological methods to determine gaseous emissions to the atmosphere are advantageous because they do not interfere with the processes of emissions and they integrate emissions over larger areas (Harper, 2005; Fowler et al., 2001) Successfully applied to crops (Denmead et al., 1978; Harper and Sharpe, 1995; Rana et al., 1998) and natural vegetation (Bussink et al., 1996; Denmead et al., 1974; Wyers and Erisman, 1998), they have rarely been used to characterize ammonia emissions from beef cattle feedyards (Hutchinson et al., 1982). The flux-gradient (FG) method treats turbulent flux as analogous to molecular diffusion, following Fick's law, with

$$Q_{FG} = -K_g \frac{\partial \rho_g}{\partial z} \tag{1}$$

where K_g ($m^2\ s^{-1}$) is called the eddy diffusivity (or turbulent transfer coefficient) of the gas of interest, ρ_g ($\mu g\ m^{-3}$) is the density of the gas, and z (m) is height; the differential expresses the vertical concentration gradient (Harper, 2005). Though eddy diffusivity is analogous to molecular diffusivity, it differs in that it is a characteristic of the flow, not the fluid; it's not constant, but varies with wind speed and atmospheric stability (Fowler et al., 2001; Prueger and Kustas, 2005); and because it is related to the size of turbulent eddies, it is proportional to distance from the surface (Thom, 1975). Because K_g is not readily known, it is found using the momentum balance method to determine the eddy diffusivity for momentum, K_m, which is then related to K_g. This requires profile measurements of gas concentration, wind speed, and air temperature to calculate a FG flux estimate. The FG method assumes that there is horizontal uniformity of air flow, that horizontal concentration gradients are negligible compared with vertical gradients, that variables are measured during steady state conditions, and that vertical flux is constant with height (Harper, 2005; Thom, 1975).

The FG method has been applied most commonly and successfully in agricultural situations where large fetch and uniform surfaces contribute to horizontal homogeneity and a well-developed fully adjusted layer. The FG method has also been used in more demanding situations, such as confined, intensive animal feeding operations. Hutchinson et al. (1982) used the FG method to estimate ammonia emission from a beef cattle feedyard. They estimated eddy diffusivity using the energy balance; NH_3 concentration profile was measured using acid solution ammonia scrubbers (gas washing) at eight heights from 1.52 to 8.84 m, located on the downwind edge of the feedyard. Mid-afternoon flux densities (1-h to 3-h sampling periods), which probably represent daily maxima, ranged from 0.64 to 2.37 kg NH_3-N $ha^{-1}\ h^{-1}$. They estimated that about 25% of N deposited on the feedyard surface was volatilized as ammonia. Harper et al. (2000) used the FG method to estimate ammonia emission from a swine lagoon. Gas washing, with 4-h sampling periods and measured at six heights from 0.2 to 2.7 m, was used to measure atmospheric ammonia concentration. Ammonia flux density from the lagoon, averaged over summer, winter and spring trials, was 12.06 ± 16.81 kg $NH_3\ ha^{-1}\ d^{-1}$. Laubach and Kelliher (2004) used the FG method to estimate methane emissions from the challenging case of free grazing dairy cattle in a finite source area. Error analysis of the FG method found that relative error of methane flux density ranged from 0.38 to 0.48 during one trial, and from 0.27 to 0.38 during a second trial. Greatest uncertainty was contributed by the methane concentration gradient. Flux-gradient derived flux was comparable to that determined by the integrated horizontal flux method when a higher gradient measurement was used, but FG underestimated flux when a lower height gradient was used.

Our objective was to use the flux-gradient method to estimate ammonia emissions from pens of a beef cattle feedyard typical of those found on the southern High Plains.

Materials and Methods

Location and Site Characteristics.

Research was conducted at a commercial beef cattle feedyard located in the Texas Panhandle (Figure 1). Mean occupancy of the 77-ha pens was 45,000 head, with a stocking density of 16 m^2 $head^{-1}$. Median capacity of feedyards in the region is 30,000 head. Though the terrain is relatively flat, the feedyard surface is complex, with several small buildings, thousands of meters of 1.5-m tall pen fences, electrical poles, manure mounded in centers of pens, and mobile cattle. A retention pond and manure stockpiles are located east of the pens. The semiarid climate of the region is characterized by hot summers and mild winters. Mean annual precipitation is 500 mm, with 75% falling from April through October. Potential evaporation is about 1500 mm, so that summer precipitation often rapidly evaporates. Prevailing winds are southerly to southwesterly, with wind direction almost half the time between 160° and 250°.

Six field campaigns were conducted; during summer 2002, 2003, and 2004, during winter 2003 and 2004, and during spring 2005 (Table 1). In each campaign, an instrument tower was installed in a pen

Table 1. Dates and meteorological conditions during field campaigns at the beef cattle feedyard.

Campaign	Date	Max. air temp.	Mean. air temp.	Min. air temp.	Mean relative humid.	Mean wind speed	Total precip.
		C	C	C	%	$m\ s^{-1}$	mm
Summer 02	19Aug-24Aug	35	25	19	68	4.7	11.7
Summer 03	14Jul-31Jul	42	28	16	21	2.6	0
Summer 04	14Jun-6Jul	37	23	14	70	5.4	69.7
Winter 03	15Jan-24Jan	24	1.4	-8.8	63	2.4	0
Winter 04	29Jan-9Feb	19	2.1	-8.9	68	3.5	tr
Spring 05	28Mar-12Apr	28.1	11.8	0	48	3.9	64

without cattle in a location intended to maximize upwind fetch in the direction of expected prevailing winds (Figure 1). In Summer 02, a 6-m tower was centered on the north margin of the pen area; in Winter 03 a 6-m tower was near the center of the pen area; and in Summer 03, Winter 04, Summer 04 and Spring 05, a 10-m tall instrument tower was erected near the center of the northeast quadrant of the pen area.

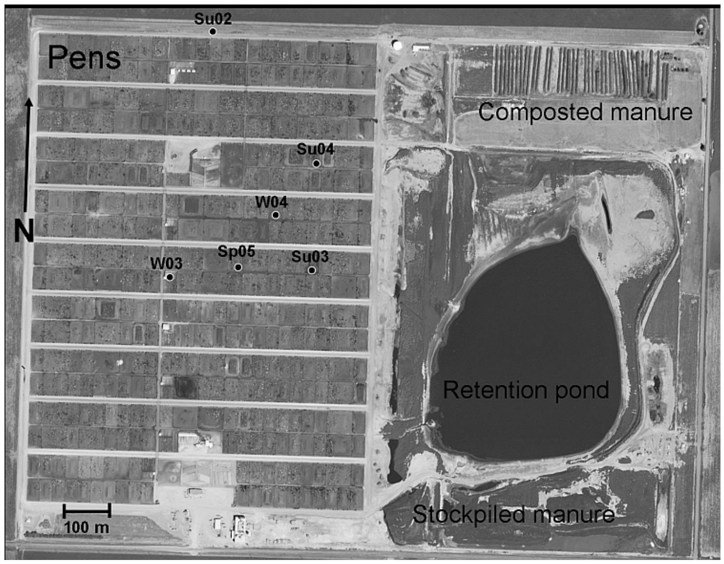

Figure 1. Layout of commercial feedyard, with locations of instrument tower during the six field campaigns.

Atmospheric Ammonia Concentration.

Ammonia concentration was measured using acid gas washing in Summer 02, Winter 03, Summer 03, Winter 04 and Spring 05 (Table 2). Ammonia was trapped in gas washing bottles by first drawing air through a teflon filter to remove particulates, then bubbling it through an impinger in 80 to 120 ml of 0.1 N H_2SO_4. Air flow rate of each gas washing bottle was measured with a precision, calibrated flow meter (Dry Cal DC Lite, Bios International, Butler, N; mention of trade or manufacturer names is made for information only and does not imply endorsement, recommendation, or exclusion my USDA-ARS.) at the beginning and

Table 2. Details, by campaign, for measurement of atmospheric ammonia concentration.

Campaign	Sampling heights	Ammonia method†	Sampling period, daytime	Sampling period, nighttime
	m		h	h

Summer 02	1.5, 3, 6	GW	3	12
Summer 03	2, 4, 6, 8	GW	3	3 or 9
Summer 04	3, 6	CH	0.33	0.33
Winter 03	2, 3, 4, 5, 6	GW	4	16
Winter 04	2, 4, 6, 8	GW	2	2
Spring 05	3, 4, 5, 6	GW	2	2

† GW is acid gas washing; CH is chemiluminescence.

end of each sampling period. Nominal air flow rate was 6 L min^{-1}. At the beginning of a sampling period, gas washing bottles with fresh acid were sealed and transported to the tower, exchanged with the bottles there, and sealed bottles with samples were returned to the laboratory, where each sample was diluted to 100 ml with acid, 30 ml was decanted into a sample bottle, and then all samples were refrigerated until analysis. A calibrated flow injection analyzer (QuickChem FIA+ 8000, Lachat Instruments, Milwaukee, WI.) was used to quantify ammonium in the samples, with a minimum detection limit of about 10 µg L^{-1}. This corresponded to atmospheric ammonia concentrations of less than 1 µg m^{-3}. However, experience indicated that the minimum detection limit of atmospheric ammonia was probably closer to 5 µg m^{-3}.

In Summer 04, ammonia concentration was measured continuously using a chemiluminescence analyzer (17C, Thermo Environmental Instruments, Franklin, MA). Ammonia concentration at 3 and 6 m was measured sequentially using a 3-way solenoid that switched gas sampling lines from one height to the other every 10 minutes. Due to the response time of the analyzer, only data from the last 3 minutes of each 10 minutes were averaged. See Baek et al. (2006) for further details.

Profiles of wind speed and air temperature were defined at the same heights as atmospheric ammonia concentration. Cup anemometers (12102M, R.M. Young, Traverse City, MI) measured wind speed and aspirated, fine-wire (25.4 µm diameter) thermocouples (ASPTC, Campbell Scientific, Logan, UT) measured air temperature. Other meteorological measurements included relative humidity and air temperature (HMP45, Vaisala, Helsinki, Finland), wind direction (12005, R.M. Young, Traverse City, MI) and precipitation (TE525, Campbell Scientific, Logan, UT). Outputs from meteorological instruments were automatically recorded to a data logger (CR23X, Campbell Scientific, Logan, UT) that sampled instruments every 5 s and calculated 1-min means.

Flux-Gradient Estimates of Ammonia Flux.

Ammonia flux was estimated from measured profiles of ammonia, wind speed and air temperature using

$$Q_{FG} = \frac{-k^2}{F} \frac{du}{d\left[\ln\left(z - d_0\right)\right]} \frac{dA}{d\left[\ln\left(z - d_0\right)\right]} \tag{2}$$

(Thom, 1975) where k is von Karman's constant, d_0 is zero plane displacement, and the derivatives represent the slopes of the regressions of wind speed (u, m s^{-1}) and ammonia concentration (A, µg m^{-3}) against the log of height (z-d_0); F is a factor to correct for non-neutral atmospheric stability (Thom, 1975; Flesch et al., 2002) and is discussed later. Using a subset of data when stability was near-neutral, displacement height, d_0, was calculated by finding the d_0 that maximized the r^2 of the log-linear regression of ln(z-d_0) against wind speed. Mean d_0 was 0.39 m. The slope of the wind speed profile will always be positive. The log-linear slope of the ammonia profile will be negative when the surface is emitting ammonia and positive when the surface is absorbing ammonia.

Stability Corrections to Flux Estimates.

When buoyancy forces are present, a correction for thermal stability is required in Eq.(2). In unstable conditions, with a lapse temperature profile, flux is enhanced by thermal buoyancy. In stable conditions, with an inverted temperature profile, flux is dampened by thermal buoyancy. Thermal effects are accounted for by first quantifying stability and then applying semi-empirical relationships that are functions of stability (Prueger and Kustas, 2005). The gradient Richardson number stability parameter was calculated using

$$R_g = \frac{g}{T} \frac{(z_g - d_0)\dfrac{\partial T}{\partial\left[\ln\left(z_g - d_0\right)\right]}}{\left\{\dfrac{\partial u}{\partial\left[\ln\left(z_g - d_0\right)\right]}\right\}^2} \tag{3}$$

where g = 9.81 m s^{-2} is acceleration of gravity, $\overline{T}$ (K) is mean air temperature between two heights, and $z_g = \sqrt{z_i z_j}$ is the geometric mean height between heights *i* and *j*. The Monin-Obukhov stability parameter, ζ, was calculated as a function of R_g using

$$\zeta = 0.67 R_g, \qquad R_g < 0 \tag{4}$$

$$\zeta = R_g (1 - 5R_g), \qquad R_g > 0 \tag{5}$$

(Högström, 1996). Stability expressions were those of Dyer (1974) modified by Högström (1988) to correspond to a von Karman's constant k = 0.4:

$$\phi_m = (1 - 15.2\zeta)^{-0.25}, \qquad \zeta < 0 \tag{6}$$

$$\phi_m = 1 + 4.8\zeta, \qquad \zeta > 0 \tag{7}.$$

One source of uncertainty in the FG approach is the relationship between the eddy diffusivities. Although it is often assumed that the eddy diffusivities for momentum, heat or mass are equal, Flesch et al. (2002) pointed out that this is not usually the case. This inequality is embodied in the Schmidt number:

$$S_c = \frac{K_m}{K_g} = \frac{\phi_c}{\phi_m} \tag{8}$$

where K_m (m^2 s^{-1}) is the eddy diffusivity for momentum, and ϕ_c and ϕ_m are the Monin-Obukhov dimensionless parameters for the ammonia gradient and wind shear, respectively. Flesch et al. (2002) found experimentally that S_c = 0.63 ± 0.31. They found that the FG method underestimated emission rate, compared with that determined by the integrated horizontal flux method, on average by 30% because the effective Schmidt number, based on commonly employed calculations of ϕ_g and ϕ_m, was greater than the experimentally determined value of S_c = 0.63. We applied their recommendation to correct the FG equation with

$$F = \left(S_c\, \phi_m^2\right) \tag{9}.$$

Data were screened by wind direction to ensure adequate upwind fetch and to eliminate observations of ammonia concentration that were possibly affected by sources other than feedyard pens, such as the retention pond or manure stockpiles. Mean daily flux was calculated by time weighted averaging of sampling period flux estimates. An exception was in Winter 2004 and Spring 2005, when ammonia concentration was measured during 2-h sampling periods every 3 h during daytime, and one 2-h sample was taken each night. Daily flux was integrated using the trapezoidal rule.

Results and Discussion

Ammonnia flux densities

Mean daily NH_3 flux density was highly variable from campaign to campaign (Table 3). Summer flux ranged from 43 to 125 µg m^{-2} s^{-1} and averaged 72 µg m^{-2} s^{-1}. Greater variability observed in Summer 04 was because of more frequent precipitation. Flux was suppressed immediately following rain and increased as feedyard pens dried. Greatest flux was observed during Summer 03, when environmental conditions were hot and dry (Table 1). Mean daily winter flux density (39 µg m^{-2} s^{-1}) averaged about half that of summer, and ranged from 10 to 78 µg m^{-2} s^{-1}. Lowest winter fluxes were observed on coldest days, though even when air temperatures were below freezing, fluxes ranged from 6 to 26 µg m^{-2} s^{-1}. Urine, the primary source of ammonia leaves cattle at 38 C, so even though it was cold, hydrolysis of urea occurred and ammonia was volatilized. Spring flux densities averaged greater than those in some summer campaigns. Mason (2004) showed greater ammonium content of feedyard pen manure during spring, and this greater availability of ammonia may have contributed to the spring fluxes. Greater day to day variability was also observed during the spring campaign, with fluxes ranging from 31 to 151 µg m^{-2} s^{-1}. Ammonia concentrations greater than 1000 µg m^{-3} and wind speeds greater than 12 m s^{-1} during some sampling periods contributed to the large fluxes observed on DOY 88 of Spring 05. Greater precipitation (69.7 mm, Table 1) in Summer 04 resulted in greater variability in daily ammonia emissions, with the coefficient of variation twice that observed in Summer 03, when there was no precipitation during the study period. Cooler temperatures and wetter conditions during Summer 02 and Summer 04 contributed to lower emissions compared with Summer 03.

Ammonia emission rates and emission factors

Ammonia-N loss averaged 3930 kg d^{-1} during summer and 2150 kg d^{-1} during winter (Table 3). Assuming that the average of summer and winter NH_3-N emission rates (3040 kg d^{-1}) was representative of the mean daily emission rate throughout the year, and that the annual production of the feedyard was 100,465 head (2.25 turnovers yr^{-1}), gives an emission factor of 11.0 kg NH_3-N $head^{-1}$ yr^{-1}. USEPA (2004) assigned an emission factor of 11.4 kg NH_3-N $head^{-1}$ yr^{-1} for beef cattle in drylots. Todd et al. (2007) reported mean ammonia-N emissions of 6110 and 2740 kg d^{-1} in summer and winter, respectively, using the same ammonia concentration data and an inverse dispersion model to estimate emissions. The annualized emission factor for feedyard pens was 15.9 kg NH_3-N $head^{-1}$ yr^{-1}, or 44% greater than the emission factor derived from the flux-gradient emission estimates from this study.

Ammonia loss and fed nitrogen

Nitrogen in rations fed to cattle ranged from 6900 to 10650 kg d^{-1}. The formulation of the diet was changed in April 2003, with crude protein increasing from 13.5% to 14.5%, and is seen as an increase in N fed from 0.16 to 0.19 kg $head^{-1}$ d^{-1} (Table 3). Optimal crude protein in beef cattle diets is about 13% (Gleghorn et al., 2004), so the new diet provided excess N, which is subsequently excreted in the urine as urea. This increase in excreted N could partly account for the increase in ammonia emission seen from Summer 02 to Summer 03 and from Winter 03 to Winter 04. Ammonia-N loss ranged from 30 to 66% of fed N in summer, and averaged 45%. In winter, NH_3-N loss was from 17 to 37% of fed N and averaged 27%. Springtime loss was 43% of fed N. Annualized NH_3-N loss was 36% of fed N. In an independent study in 2003 and 2004 at the same feedyard, Harper et al. (2004) used open path lasers to measure ammonia concentration and an inverse dispersion model to estimate flux. They found that NH_3-N loss was 53% and 29% of fed N in summer and winter, respectively. Flesch et al. (2007) found that 63% of fed N was lost as ammonia-N during summer 2004 at the same feedyard. Todd et al. (2007), using the same ammonia concentration data as this study and an inverse dispersion model reported that ammonia-N loss was 68% of fed N in summer and 36% in winter, with an annualized mean of 53%. Erikson and Klopfenstein (2001) used a nitrogen balance method and estimated that in Nebraska 60-70% of excreted N was lost as gaseous N during summer, and 40% during winter-spring. This was 53-63% of fed N during summer and 35% during winter-spring.

Table 3. Mean daily ammonia flux densities, ammonia-N emissions, and fed nitrogen. Coefficient of variation (CV) is for the mean of the mean daily flux.

Campaign	No. of days	Mean daily NH_3 flux	Mean daily NH_3-N emission rate	CV	N fed	NH_3-N lost as fraction of fed N
		$\mu g\ m^{-2}\ s^{-1}$	kg d^{-1}	%	kg $head^{-1}$ d^{-1}	%
Summer 02	4	48	2610	15	0.16	38
Summer 03	7	108	5960	12	0.19	66
Summer 04	7	59	3230	24	0.22	30
Winter 03	5	21	1150	36	0.16	17
Winter 04	6	57	3140	34	0.20	37
Spring 05	4	79	4350	71	0.20	43

Flux-gradient method and inverse dispersion model compared

Ammonia-N emission rates from this study estimated using FG averaged 22% less in winter and 36% less in summer than emissions quantified using an inverse dispersion model (Todd et al., 2007)(Table 4). Annualized ammonia-N emission rate using the FG method was 3040 kg d^{-1}, compared with 4430 kg d^{-1} estimated using the inverse dispersion model, a 31% reduction.

Table 4. Averaged ammonia-N emission rate estimates from an inverse dispersion model (Todd et al., 2007) and the flux-gradient method (this study) compared. Days included in each mean do not exactly correspond.

Season	Inverse dispersion model	Flux-gradient method
	kg NH_3-N d^{-1}	
Winter	2740	2150
Summer	6110	3930
Annualized	4430	3040

Wilson et al. (2001) compared three methods, flux-gradient (FG), integrated horizontal flux (IHF), and inverse dispersion model (BLS) to estimate trace gas flux in disturbed wind flow such as found in feedyards or lagoons. Disturbed flow was defined as advective flow in which the assumption of horizontal homogeneity may be invalid. The IHF method is a mass balance approach that is assumption free, in which the horizontal flux across a vertical plane is integrated from the surface to the top of the trace gas plume to derive the vertical flux. The FG flux estimates were consistently less than the IHF or BLS estimates, and Wilson et al. (2001) concluded that the latter two methods were preferable to FG in disturbed flow conditions. Laubach and Kelliher (2004) compared FG and IHF estimates of methane flux from a finite source area of grazing dairy cattle. They found that the Schmidt number (S_c) was a function of the ratio of measurement height to source area distance, ranging from 0.7 to 1.2, which limited the effectiveness of FG estimates compared with the IHF method. They also recommended the IHF method over FG because the former did not rely on similarity assumptions and had smaller measurement error. Flesch et al. (2002) found that FG flux estimates averaged 30% less than those of the IHF method, again because of variability and uncertainty in S_c. Our results similarly show that the FG method yields lower flux estimates than an inverse dispersion model, with the reduction in the same range as the above discussed comparisons.

Conclusions

Ammonia-N emissions from a typical commercial beef cattle feedyard on the southern High Plains estimated using the flux-gradient method were 45% of fed nitrogen in summer and 27% of fed nitrogen in winter, and on an annual basis, averaged 36% of fed nitrogen. Winter ammonia emissions were about half those in summer. Year to year variability was partly explained by differences in temperature and precipitation, and an increase in fed nitrogen and subsequent increase in ammonia emission. This suggests that optimizing fed nitrogen through practices such as phase feeding could help minimize ammonia emissions. Ammonia emissions estimated using the flux-gradient method were 22 to 36% less than those derived from an inverse dispersion model. This is consistent with results of other studies that showed the tendency of the flux-gradient method to give lower emissions in conditions of disturbed flow. Uncertainty of the Schmidt number and violation of the assumption of homogeneous flow were possible contributors to the lower emission estimates of the flux-gradient method. Longer term monitoring of ammonia emissions is needed to better define annual variability, emission rates and emission factors.

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